High-T$_c$ Superconductors with Antiferromagnetic Order: Limitations on Spin-Fluctuation Pairing Mechanism

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The very intriguing antagonistic interplay of antiferromagnetism (AF) and superconductivity (SC), recently discovered in high-temperature superconductors, is studied in the framework of a microscopic theory. We explain the surprisingly large increase of the magnetic Bragg peak intensity I_Q at $Q \sim (\pi, \pi)$ in the magnetic field $H \ll H_{c2}$ at low temperatures $0 < T \ll T_c, T_{AF}$ in $La_{2-x}Sr_xCuO_4$. Good agreement with experimental results is found. The theory predicts large anisotropy of the relative intensity $R_Q(H) = (I_Q(H) - I_Q(0))/I_Q(0)$, i.e. $R_Q(H \parallel c - axis) \gg R_Q(H \perp c - axis)$. The quantum (T=0) phase diagram at H=0 is constructed. The theory also predicts: (i) the magnetic field induced AF order in the SC state; (ii) small value for the spin-fluctuation coupling constant $g < (0.025 - 0.05) \ eV$. The latter gives very small SC critical temperature $T_c(\ll 40 \ K)$, thus questioning the spin-fluctuation mechanism of pairing in HTS oxides.

Introduction - The antagonistic interplay of superconductivity (SC) and antiferromagnetic (AF) order has been intensively studied in compounds with regular distribution of magnetic ions, for instance in $RERh_4B_4$, $REMo_6S_8(Se_8)$, etc. - the systems with localized rare earth (RE) magnetic moments, and UPt_3, URu_2Si_2 UCu_5, UPd_2Al_3 etc. - the systems with itinerant moments [1]. The interest in the coexistence problem of the AF and SC order has increased after the discovery of high-temperature superconductors (HTS), in which the antiferromagnetic fluctuations and the tendency to the AF order are pronounced. This fact implies inevitably several important questions: (i) do the AF and SC order coexist in these materials and how they influence each other? (ii) if the spin fluctuations are responsible for pairing in HTS oxides, in which way are they affected by the AF order? (iii) is the AF order compatible with other pairing mechanisms, such as the phonon one [2]? There is a strong believe that by answering these questions an important step will be done towards the understanding of the pairing mechanism in HTS oxides, which is still under debate.

The first experimental evidence for the coexistence of the AF and SC order in $La_{2-x}Sr_xCuO_4$ (for $x < 0.15$) was reported in Ref.([3]), which gave an impetus for theoretical studies of this problem in HTS oxides [4]. In the last few years careful neutron magnetic scattering measurements on La_2CuO_{4+y} [5] , $La_{2-x}Sr_xCuO_4$ (for $x = 0.12$, $T_c = 12 \ K$, $T_{AF} \approx 25 \ K$) [6], $YBa_2Cu_3O_{6.5}$ (with $T_c = 55 \ K$, $T_{AF} \approx 310 \ K$) [8], $YBa_2(Cu_{1-y}Co_y)_3O_{7+\delta}$ (with $y = 0.013$ and $T_c = 93 \ K$, $T_{AF} \approx 330 \ K$) [9], have given sufficient evidence for the coexistence of the AF and SC order. The existence of the pronounced elastic magnetic Bragg peak at the wave vector $\mathbf{Q} \sim \mathbf{Q}_{AF} = (\pi, \pi)$ means that in these HTS oxides static AF correlations are developed on the length scale $\xi_m \gtrsim 400$ Å [5], [6]. Since ξ_m is much larger than the SC coherence

length in the a-b plane, $\xi_{ab} \sim (20 - 25)$ Å, this means that these static AF correlations can be considered as a static AF order which competes with superconductivity far outside the vortex core. The experiments demonstrated that the effective magnetic moment μ is small, i.e. $\mu \lesssim 0.1 \ \mu_B$ in $La_{2-x}Sr_xCuO_4$ and $\mu < 0.05 \ \mu_B$ in $YBa_2(Cu_{1-y}Co_y)_3O_{7+\delta}$.

The magnetic neutron scattering experiments performed on $La_{2-x}Sr_xCuO_4$ ($x = 0.12$, $T_c = 12 \ K$, $T_{AF} \approx 25$ K [6]) in magnetic field $H \sim 10 \ T$ and at $T = 4 \ K$ show surprisingly large increase of the elastic magnetic Bragg peak intensity $I_{\mathbf{Q}}$ by as much as 50 % of that at $0 \ T$. This behaviour of $I_{\mathbf{Q}}(H)$ was confirmed recently on the same compound [7] ($T_c = 29 \ K$, $T_{AF} \approx 25$ K), where $I_{\mathbf{Q}}(H) - I_{\mathbf{Q}}(0) \sim \alpha H \ln bH$ with an increase of $I_{\mathbf{Q}}(H)$ as much as 100% at low T. Such a large increase of $I_{\mathbf{Q}}(H)$ (and its peculiar field dependence) exceeds by far the formally similar effect in the magnetic superconductor $HoMo_6S_8$ [10] which exhibits a sinusoidal magnetic order with $Q \ll Q_{AF}$ where $I_{\mathbf{Q}}(H) - I_{\mathbf{Q}}(0) \sim H^2$ for $H \lesssim 200 \ Oe$. The latter is due to the suppression of the SC order in the presence of the exchange (or spin-orbit) scattering, as it was explained in [11].

The problem of the coexistence of the AF and SC order in high magnetic fields was recently studied in the framework of the phenomenological Ginzburg-Landau (GL) theory [12] by assuming that the SC and AF order parameters are small ($\mid \psi \mid \ll 1$, $m_Q \ll 1$). The logarithmic behavior of $I_{\mathbf{Q}}(H)$ is found to be due to the suppression of SC in the vortex state. However, the pronounced increase of $I_{\mathbf{Q}}(H)$ in the experiments on $La_{2-x}Sr_xCuO_4$ is realized at very low temperatures ($T \ll T_c$) and in low fields $H \ll H_{c2}$, where $H_{c2}(T \ll T_c) \approx (50 - 80) \ T$. Thus the assumption that the SC order parameter is small ($\mid \psi \mid \ll 1$) is not valid in these experiments and a *microscopic theory* of the problem is needed. The latter is proposed and elaborated in this paper. The fact that

$\mu(\sim 0.1-0.05)$ is small in the AFS systems of HTS oxides points to a *weak itinerant antiferromagnetism*, which we also assume in the following - the WIAF model. We shall calculate in this model the intensity $I_Q(\mathbf{H})$ in the coexistence phase for various field orientations. The quantum $(T = 0\ K)$ phase diagram is constructed and the condition for the magnetic field induced AF order in the SC state is found.

WIAF model for AFS at $T \ll T_c$ - The interplay of the AF and SC order is studied in the framework of the weak coupling Hamiltonian

$$\hat{H} = \hat{H}_0(\vec{p} - \frac{e}{c}\mathbf{A}) + \hat{H}_Z + \sum_i gn_{i\uparrow}n_{i\downarrow} + \hat{H}_{BCS}, \quad (1)$$

where $\hat{H}_Z$ describes the Zeeman coupling of spins with $\mathbf{H}$ and $\mathbf{A}$ is the vector potential. The Hubbard term ($\sim g$) is responsible for the weak itinerant AF order, while the $\hat{H}_{BCS}$ describes superconductivity with the SC order parameter $\Delta(\mathbf{k}, \mathbf{R}) = Y_d(\mathbf{k})\Delta_0(\mathbf{R})$, where $\mathbf{R}$ is the mass centrum and $Y_d(\mathbf{k})$ is the d-wave function.

The study of the orbital effect of the magnetic field is mathematically complicated since in fields $H_{c1} < H < H_{c2}$ the vortex state is realized and $\Delta_0(\mathbf{R})$ varies in space. This variation occurs on a much larger scale ξ_{ab} than the atomic length a (the length scale of the AF order), i.e. $\xi_{ab}, d_v \gg a$. The HTS materials are extreme type II superconductors with the GL parameter $\kappa \gg 1$, and since the AF order is developed on the large scale $\xi_m \gg \xi_{ab}$ it is plausible to approximate $\Delta_0(\mathbf{R})$ by its average value over the unit vortex cell with the intervortex distance d_v, i.e. we put $\Delta_0(\mathbf{R}) \to \sqrt{< \Delta_0^2(\mathbf{R}) >_v}$ similarly as it has been done in Ref. [12]. In the case when $H \ll H_{c2}$ one has [12] $< \Delta_0^2(\mathbf{R}) >_v \approx \Delta_0^2 \varphi^2(h)$ with $\varphi(h) \approx [1 - (h/2)\ln 3/h]^{1/2}$ (for triangular vortex lattice) and $h = H/H_{c2}(\theta)$. The upper critical field $H_{c2}(\theta) = H_{c2}^c/(\cos^2\theta + \varepsilon^2\sin^2\theta)^{1/2}$ depends on the angle θ between $\mathbf{H}$ and the crystal $c-axis$ where $\varepsilon = H_{c2}^c/H_{c2}^{ab}$. So, the (orbital) effect of the vortex lattice on superconductivity is accounted for by multiplying the SC order parameter by $\varphi(h)$. For further purposes we define the SC order parameter $\psi(h) = \Delta(h)/\Delta_0\varphi(h)$, where Δ_0 is the SC gap in absence of the AF order and at $h = 0$. We shall see that this physically plausible approximation gives very good description of experiments on $La_{2-x}Sr_xCuO_4$.

Free-energy of AFS - The magnetism in the WIAF model at $T \ll T_{AF}$ is treated in the Hartree-Fock approximation [13]. By assuming that: (i) $h_{ex}(\equiv gm_Q) \ll \Delta$, which is realized in $La_{2-x}Sr_xCuO_4$ - see below, (ii) $t = T/T_c \ll 1$ (iii) $\chi_{sQ}/\chi_{nQ}(\Delta_0) \lesssim 1$ - see below, the normalized Gibbs free-energy $\tilde{G}(\equiv \chi_{n0}G)$ functional for the AFS in magnetic field $\mathbf{h}$ has the form

$$\tilde{G} = -\frac{a_s}{4}\varphi^2(h)\psi^2\ln\frac{e}{\psi^2} + \frac{a}{2}m^2 + \frac{b}{4}m^4 - \mathbf{m}\mathbf{h}\tilde{h}_{c2}$$

$$-\frac{a_Q}{2}m_Q^2 + \frac{b_Q}{4}m_Q^4 + \frac{3}{4}bm^2m_Q^2 + c\psi m^2 + c_Q\psi m_Q^2. \quad (2)$$

The first term describes the superconducting condensation energy (in absence of the AF order) with $a_s = (N(0)\Delta_0)^2$. The second and third terms are due to the induced ferromagnetic (F) moment m in the magnetic field, where $a = 1 - 2g\chi_{no} > 0$ and $\chi_{no} = N(0)/2$ is the free-electron normal state spin susceptibility at $\mathbf{q} = \mathbf{0}$. The parameter b depends on the quasiparticle spectrum and for a weakly anisotropic spectrum one has [13] $b = [(N'(0)/N^2(0))^2 - N''(0)/3N^3(0)]/2$, where $N(0)$, $N'(0)$, $N''(0)$ are the density of states and their derivatives at the Fermi surface, respectively. The fourth term is the energy of the magnetic moment in the field $\mathbf{h} = \mathbf{H}/H_{c2}$ where $\tilde{h}_{c2} = N(0)(\mu_B H_{c2})$. The weak itinerant AF order (in absence of the SC order) is described by the first two terms in the second line in $Eq.(2)$, where m_Q is the AF order parameter, $a_Q = (2\chi_{nQ}g - 1)\chi_{no}/\chi_{nQ}$ and χ_{nQ} is the free-electron normal state spin susceptibility at $\mathbf{Q} \sim \mathbf{Q}_{AF} = (\pi, \pi)$, while $b_Q = 3b/8$ as in [11]. The third term in the second line in $Eq.(2)$ is due to the interaction of the F and AF components of the magnetic moment, while the terms proportional to $c \approx \varphi(h)(\chi_{n0}/\chi_{s0}(\Delta_0)/2$ and $c_Q \approx r\varphi(h)(\chi_{nQ}/\chi_{sQ}(\Delta_0) - 1)/2$ are due to the competition of the F and SC order, and of the AF and SC order, respectively. Here, $r = \chi_{n0}/\chi_{nQ}$ where $\chi_{s0}(\Delta_0)$ and $\chi_{sQ}(\Delta_0)$ are the free-electron spin susceptibility in the SC state with $\psi = 1$ at the wave vectors $\mathbf{0}$ and $\mathbf{Q}$, respectively. In the case of a clean ($l \gg \xi_{ab}$) singlet d-wave superconductor, which we assume here, χ_{s0} is strongly suppressed, i.e. $\chi_{s0}/\chi_{n0} \approx 1.4(T/\Delta) \ll 1$ at $T \ll T_c$. The susceptibility χ_{sQ} at $Q \approx (\pi, \pi)$ is much less suppressed than at $q = 0$, i.e. $\chi_{sQ}(\Delta_0) \approx \chi_{nQ}(1 - \Delta_0/E_Q)$ for $\Delta_0/E_Q \ll 1$, where E_Q depends on the electronic spectrum. (For a parabolic spectrum $E_Q \approx \hbar v_F Q \gg \Delta$.). We stress, that in the proposed microscopic theory the term in the Gibbs free-energy which describes the interaction of the AF and SC order has the form $\tilde{G}_{int}^{micro} \sim |\psi| m_Q^2$ at $T \ll T_c$, contrary to the GL expression $\tilde{G}_{int}^{GL} \sim |\psi|^2 m_Q^2$ which holds at $T \lesssim T_c$ and which is assumed in Ref. [12].

Phase diagram at $h = 0$ - By minimizing $\tilde{G}(\psi, m_Q, m; h = 0, T = 0)$ the quantum phase diagram - shown in $Fig.1$, is calculated by taking physically plausible values for $c_Q = 0.01$, $b_Q = 1/6$. The latter values can also explain $I_Q(\mathbf{H})$ for $H \neq 0$ - see below. The phase diagram is studied in terms of the parameters $a_s(\equiv 2.4 \times 10^{-3}\ A_s)$ and $a_Q(\equiv (4/e) \times 10^{-2}\ A_Q)$ which contains the following transition lines: (i) a *first order* transition line AF$\longleftrightarrow$SC given by $A_Q = (e/2)(A_s)^{1/2}$ for $A_s < 1$; (ii) a *first order* transition line AFSC$\longleftrightarrow$AF described by $A_Q = (\sqrt{e}/2)A_s\exp(1/2A_s)$ for $A_s > 1$ and (iii) a AFSC$\longleftrightarrow$SC *second order* line $A_Q = e/2$ on which the AF order starts growing from zero.

Magnetic Bragg peak intensity $I_Q(h)$ - Since $I_Q(h) \sim m_Q^2$ we first calculate m_Q^2 in the case when $\mathbf{H} \parallel c-axis$ ($\mathbf{m}_Q \perp c-axis$) by taking $N(0) \approx 1/100\ meV$ [15], $\Delta_0 \approx 10\ meV$ (in $La_{2-x}Sr_xCuO_4$) which gives $a_s \approx 10^{-2}$. Furthermore, in clean d-wave SC one has $c \gtrsim 10$ at low $T \lesssim 1\ K$, while $b \approx 0.5$ follows from the WIAF
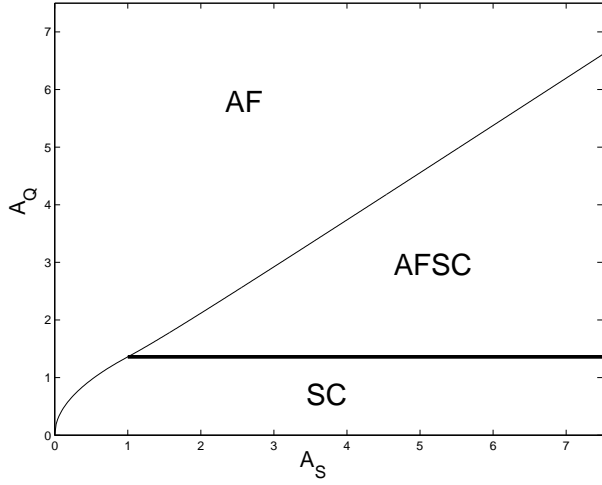
FIG. 1: Quantum phase diagram (A_s, A_Q) of the AFS system for $c_Q = 0.01$ and $b_Q = 1/6$. The thin and thick lines are the first and second order transition lines, respectively. *AF* - $(m_Q \neq 0, \psi = 0)$, *SC* - $(m_Q = 0, \psi \neq 0)$ and *AFSC* - $(m_Q \neq 0, \psi \neq 0)$.
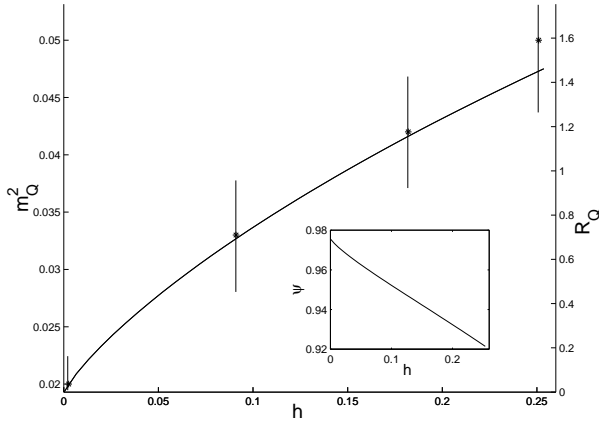

FIG. 2: Case $H \parallel c-axis$. Magnetic field ($h = H/H_{c2}^c$) dependence of m_Q^2, R_Q and ψ (inset) for $a_s = 0.01$, $a_Q = 0.028$ $b = 0.5$, $t = 0.1$, $r = 0.25$, $\beta = 0.1$ and $\tilde{h}_{c2} = 0.1$. Points and errorbars are experimental results [7] by assuming that $H_{c2}^c \approx 50~T$.

theory [13]. We take $r \approx 0.25$ in accordance with the model calculations of the quasiparticle spectrum in HTS oxides [14]. Since in low-T_c AFS systems one has $\beta \ll 1$ we take $\beta \approx 0.1$, which is also in accordance with the model calculations [14] for $\chi_{nQ}^0 - \chi_{sQ}^0$.

The calculated functions $m_Q^2(h)$, $R_Q(h)$ and $\psi(h)$, for $a_s = 0.01$, $b = 0.5$, $t = 0.1$, $r = 0.25$, $\beta = 0.1$, $a_Q = 0.028$, are shown in $Fig.2$. The relative intensity $R_Q(h)$ behaves like $R_Q(h) \approx A(1 - \varphi(h))$, while the quadratic term $\sim Bh^2 < 0.01$ (which dominates in $HoMo_6S_8$) is negligible. By assuming $H_{c2}^c \approx 50~T$ (at $T \ll T_c$), and having in mind the large experimental errorbar, one obtains very good *quantitative agreement* of the proposed theory and experimental results [7] on
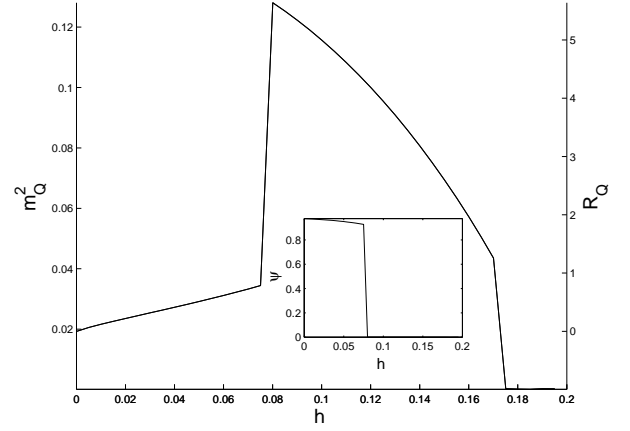

FIG. 3: Case $H \perp c-axis$. Magnetic field ($h = H/H_{c2}^{ab}$) dependence of m_Q^2, R_Q and ψ (inset) for parameters in $Fig.2$ and for $h_{c2}^{ab} = 10h_{c2}^c$.

$La_{2-x}Sr_xCuO_4$, as it is seen in $Fig.2$. Note, that in this compound (and for $h = 0$) the bare AF order parameter m_{Q0} is drastically suppressed by superconductivity, i.e. $m_Q^2 = m_{Q0}^2 - 2c_Q\psi \ll m_{Q0}^2$. We stress also that the AF order is not only realized inside the vortex core ($0 < R_{cor} < \xi_{ab}$) but also in the bulk region given by $\xi_{ab} < R_{bulk} < d_v$ ($d_v \gg \xi_{ab}$ at $H \ll H_{c2}$), which is in accordance with the experiments on $La_{2-x}Sr_xCuO_4$ [6]. The calculations also show that in the narrow region $h = 0.3 - 0.4$ there is a steep decrease of ψ while m_Q grows sharply (similarly as in $Fig.4$ for $\mathbf{H} \perp \mathbf{c}$-axis).

Second, in the field $\mathbf{H} \perp c$-axis and for $H_{c2}^{ab}(T = 1~K) \sim 10H_{c2}^c(T = 1~K)$ one gets a small value for $R_Q(h) \sim 0.01$ at $H \sim 10~T$, while for $h = 0.07 - 0.08$, m_Q^2 increases sharply and ψ drops to zero - $Fig.3$. Due to this large field anisotropy one expects that in poly-crystalline samples of $La_{2-x}Sr_xCuO_4$ superconductivity vanishes in fields $20~T < H < 40~T$ in the percolation process [16].

A very interesting situation is realized if $c_Q(\ll 1)$ is increased by 20% in which case the AF order is *induced* ($m_Q \neq 0$) in magnetic fields $h > 0.07$ - see $Fig.4$. This sensitivity of the AF order on c_Q may explain its absence in some HTS oxides.

Discussion - The microscopic theory proposed here predicts that the interplay of the SC and AF order is controlled by the interaction parameter c_Q. The experiments on $YBa_2Cu_3O_{7-x}$ imply that $c_Q(\ll 0.01)$ is very small, and therefore the SC order coexists with the AF itinerant magnetism (with $m_Q \approx m_{Q0} \ll 1$) more easily. One expects in this case, that the AF order is weakly affected by the magnetic field, contrary to the case of $La_{2-x}Sr_xCuO_4$ discussed above.

Since the WIAF model for the AF order in HTS oxides explains the intensity $I_Q(\mathbf{h})$ very well, it is a challenge to estimate the spin-fluctuation coupling constant g entering a_Q. For $a_Q \sim 0.028$ (if $b \sim 0.5$), which fits $I_Q(\mathbf{h})$ very well, or even for $a_Q \sim 1$ (but $b > 1$), one obtains

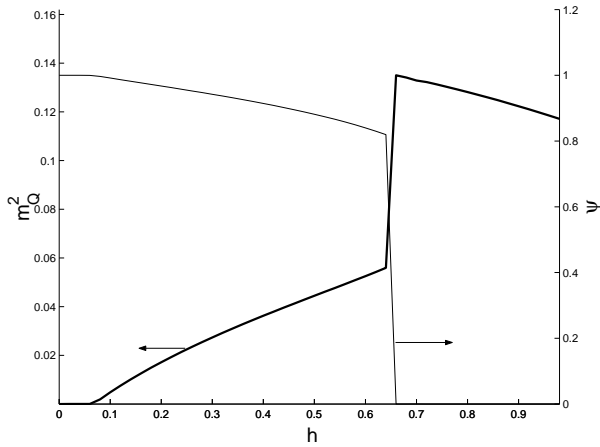FIG. 4: Induced AF order for $H \parallel c - axis$. Magnetic field ($h = H/H_{c2}^c$) dependence of m_Q^2 and ψ for $a_s = 0.01$, $a_Q = 0.028$ $b = 0.5$, $t = 0.1$, $r = 0.25$, $\beta = 0.12$ and $\tilde{h}_{c2}^c = 0.1$.

$2\chi_{nQ}^0 g \lesssim (1-2)$, which gives $g < (0.25 - 0.5)/N(0)$. For $N(0) \sim (100~meV)^{-1}$ one obtains $g < (25 - 50)~meV$, i. e. $g \ll g^{sf} \approx 0.65~eV$, where g^{sf} being the (very large) coupling constant assumed in theories based on the spin-fluctuation pairing mechanism in HTS oxides in order to explain $T_c \sim 100~K$ - see [17]. The obtained small value of $g(\ll g^{sf})$ in the AFS materials of the HTS oxides tells us that T_c, which is due to the spin-fluctuation (SF) interaction, is very small, in agreement with the value $g^{sf} \approx 15~meV$ obtained recently by analyzing the resonance peak effects in the SC state of HTS oxides [18]. Serious arguments against the SF pairing mechanism were given in Ref. [2], which are based on the experimen-

tal fact that by increasing doping from $YBa_2Cu_3O_{6.92}$ to $YBa_2Cu_3O_{6.97}$, there is a large redistribution (over ω) of the SF spectral function $Im\chi(Q,\omega)$, while T_c is changed only slightly, i.e. $T_c = 91~K$ in $YBa_2Cu_3O_{6.92}$ and $T_c = 92.5~K$ in $YBa_2Cu_3O_{6.97}$. This result can be explained by invoking very small coupling between conduction electrons and spin-fluctuations, i.e. $g^{sf} \ll 0.65$ eV. Note, that the small g value gives that $h_{ex} \sim (2-5)$ meV in $La_{2-x}Sr_xCuO_4$, i.e. the condition $h_{ex} \ll \Delta_0$ used in the calculation is fulfilled.

In conclusion, the proposed microscopic theory for the interplay of the AF and SC order in HTS oxides is able to explain quantitatively the magnetic field dependence of the magnetic Bragg scattering intensity $I_Q(\mathbf{H})$ in $La_{2-x}Sr_xCuO_4$ for $x < 0.15$. A large anisotropy of the relative intensity $R_Q(\mathbf{H})$ is predicted, i.e. $R_Q(\mathbf{H} \parallel \mathbf{c}$-axis) $\gg R_Q(\mathbf{H} \perp \mathbf{c}$-axis), which implies that in polycrystalline samples of $La_{2-x}Sr_xCuO_4$ superconductivity disappears in the percolation process in fields, 20 $T < H < 40~T$ (for $H_{c2}^c \sim 50~T$). In the quantum phase diagram (for $h = 0$) one second order and two first order transition lines meet at the same point. The theory also predicts: (i) that the AF order parameter is strongly renormalized by the SC order in $La_{2-x}Sr_xCuO_4$; (ii) that in the SC state of some HTS oxides the AF order can be induced by a finite magnetic field; (iii) that the small value for the spin-fluctuation (SF) coupling constant, $g < (0.025 - 0.05)~eV$, makes the SF mechanism ineffective in producing high T_c in HTS oxides.

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